

FORM PTO-1390 (REV 12-97)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		DATE: December //, 2001	
<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>				ATTORNEY'S DOCKET NUMBER 56603US011	
				U.S. APPLICATION NO. 10/009757 (if known, see 37 C.F.R. 1.5)	
INTERNATIONAL APPLICATION NO. PCT/EP00/06556		INTERNATIONAL FILING DATE 11 July 2000		PRIORITY DATE CLAIMED 17 July 1999	
TITLE OF INVENTION PROCESS FOR THE RECOVERY OF FLUORINATED EMULSIFIERS FROM AQUEOUS PHASES					
APPLICANT(S) FOR DO/EO/US BURKARD, Georg; HINTZER, Klaus; LOEHR, Gernot					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
1.	<input checked="" type="checkbox"/>	This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.			
2.	<input type="checkbox"/>	This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.			
3.	<input checked="" type="checkbox"/>	This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).			
4.	<input checked="" type="checkbox"/>	A proper Demand for International Preliminary Examination was made by the 19 <sup>th</sup> month from the earliest claimed priority date.			
5.	<input checked="" type="checkbox"/>	A copy of the International Application as filed (35 U.S.C. 371(c)(2)).			
	<input checked="" type="checkbox"/>	is transmitted herewith (required only if not transmitted by the International Bureau).			
	<input type="checkbox"/>	has been transmitted by the International Bureau.			
	<input type="checkbox"/>	is not required, as the application was filed in the United States Receiving Office (RO/US)			
6.	<input checked="" type="checkbox"/>	A translation of the International Application into English (35 U.S.C. 371(c)(2)).			
	<input checked="" type="checkbox"/>	is attached hereto			
	<input type="checkbox"/>	Has been previously submitted under 35 U.S.C. 154(d)(4)			
7.	<input checked="" type="checkbox"/>	Amendment to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))			
	<input checked="" type="checkbox"/>	are transmitted herewith (required only if not transmitted by the International Bureau).			
	<input type="checkbox"/>	have been transmitted by the International Bureau.			
	<input type="checkbox"/>	have not been made; however, the time limit for making such amendments has NOT expired.			
	<input type="checkbox"/>	have not been made and will not be made.			
8.	<input checked="" type="checkbox"/>	A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).			
9.	<input checked="" type="checkbox"/>	An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).			
10.	<input type="checkbox"/>	A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).			
<b>Items 11. to 16. below concern other document(s) or information included:</b>					
11.	<input type="checkbox"/>	An Information Disclosure Statement under 37 CFR 1.97 and 1.98.			
12.	<input checked="" type="checkbox"/>	An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.			
13.	<input type="checkbox"/>	A FIRST preliminary amendment.			
	<input type="checkbox"/>	A SECOND or SUBSEQUENT preliminary amendment.			
14.	<input type="checkbox"/>	A substitute specification.			
15.	<input type="checkbox"/>	A change of power of attorney and/or address letter.			
16.	<input type="checkbox"/>	Other items or information:			

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Under 37 CFR 1.10, I certify that this entry of a PCT application into US National Stage is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service addressed to the Commissioner for Patents, Washington, DC 20231, on the date indicated below.

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*Carol Decaire*

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Carol Decaire

JC07 Rec'd PCT/PTO 11 DEC 2001

U.S. APPLICATION NO. (known) 37 C.F.R. 1.10/009757		INTERNATIONAL APPLICATION NO.: PCT/EP00/06556		ATTORNEY'S DOCKET NUMBER 56603US 011	
17. <input checked="" type="checkbox"/> The following fees are submitted: <b>Basic National Fee (37 CFR 1.492(a)(1)-(5)):</b>				<b>Calculations</b>	<b>PTO Use Only</b>
<input checked="" type="checkbox"/> Search Report has been prepared by the EPO or JPO ..... \$890 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) ... \$710 <input type="checkbox"/> No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445 (a)(2)) ..... \$740 <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$1,040 <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) ..... \$100					
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				\$ 890	
Surcharge of \$ 130 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
<b>Claims</b>	<b>Number Filed</b>	<b>Number Extra</b>	<b>Rate</b>		
<b>Total Claims</b>	20 -20 =	= 0	x \$ 18	\$ 0	
<b>Independent Claims</b>	1 -3=	= 0	x \$ 84	\$ 0	
MULTIPLE DEPENDENT CLAIM(S) (IF APPLICABLE)				x \$ 280	\$ 280
<b>TOTAL OF ABOVE CALCULATION =</b>				\$ 1,170	
Reduction by 1/2 for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28)				\$	
<b>SUB TOTAL =</b>				\$ 1,170	
Processing fee of \$ 130 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest priority date (37 CFR 1.492(f)).				\$	
<b>TOTAL NATIONAL FEE =</b>				\$ 1,170	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$	
<b>TOTAL FEES ENCLOSED =</b>				\$	
				<b>Amount to be refunded</b>	\$
				<b>charged</b>	\$ 1,170
a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. 13-3723 in the amount of \$ 1,170 to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3723 A duplicate copy of this sheet is enclosed <b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b>					

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**PROCESS FOR THE RECOVERY OF FLUORINATED  
EMULSIFIERS FROM AQUEOUS PHASES**

**5 Description**

The invention relates to the work-up of waste water, in particular lightly contaminated waste water, containing fluorinated emulsifiers, as employed in the polymerization of fluorinated monomers since they do not have telogenic properties. In particular, the salts, preferably the  
10 alkali metal or ammonium salts, of perfluorinated or partially fluorinated alkanecarboxylic acids or -sulfonic acids are used. These compounds are prepared by electrofluorination or by telomerization of fluorinated monomers, which is associated with considerable effort. There has therefore been no lack of attempts to recover these valuable materials from waste water.

15 US-A-5,442,097 discloses a process for the recovery of fluorinated carboxylic acids in usable form from contaminated starting materials, in which the fluorinated carboxylic acid is, if necessary, liberated from these materials in aqueous medium using a sufficiently strong acid, the fluorinated carboxylic acid is reacted with a suitable alcohol, and the ester formed is distilled off. The starting material used here can be a polymerization liquor, in particular from emulsion  
20 polymerization, in which the fluoropolymer is prepared in the form of colloidal particles with the aid of relatively large amounts of emulsifier. "Polymerization liquor" here is taken to mean the waste water produced on isolation of the fluoropolymer by coagulation (without further process steps, such as washing). This process has proven highly successful, but requires a certain concentration of fluorinated carboxylic acid in the starting material.

25 The recovery of fluorinated carboxylic acids by distillation can also be carried out in the absence of alcohols. In this process variant, the fluorocarboxylic acid is distilled off in the form of a highly concentrated azeotrope. However, this process variant is industrially disadvantageous for energetic reasons. In addition, the resultant waste water is more highly contaminated than before  
30 the treatment.

DE-A-20 44 986 discloses a process for the isolation of perfluorocarboxylic acids from dilute solution in which the dilute solution of the perfluorocarboxylic acids is brought into adsorption contact with a weakly basic anion exchanger resin, and the perfluorocarboxylic acid present in

the solution is thereby adsorbed onto the anion exchanger resin, the anion exchanger resin is eluted with an aqueous ammonia solution and the adsorbed perfluorocarboxylic acid is thus transferred into the eluent, and finally the acid is isolated from the eluate. However, complete elution requires relatively large amounts of dilute ammonia solution, and in addition this process is very time-consuming. These disadvantages are overcome by the process disclosed in US-A-4,282,162 for the elution of fluorinated emulsifier acids adsorbed onto anion exchangers, in which the elution of the adsorbed fluorinated emulsifier acid from the anion exchanger is carried out using a mixture of dilute mineral acid and an organic solvent. In this process, the use of the acid simultaneously effects regeneration of the exchanger resin.

The use of anion exchanger resins in waste water treatment on an industrial scale is essentially hindered by the presence of fluoropolymer latex particles. The latex particles are anionically stabilized and are consequently coagulated in the anion exchanger resin. The exchanger column thus becomes blocked.

This difficulty is overcome by a proposed process for the isolation of fluorinated emulsifier resins in which the finely divided solids in the waste water are stabilized using a surfactant or a surface-active substance, and subsequently the fluorinated emulsifier acids are bound to an anion exchanger resin, and the fluorinated emulsifier acids are eluted therefrom (WO-A-99/62830). In the examples, nonionic surfactants are employed in a concentration of from 100 to 400 mg/l.

A process has now been found for the recovery of fluorinated emulsifiers from an aqueous phase, where this aqueous phase, besides the emulsifier, contains small amounts of fluoropolymer particles and possibly further substances, where

- an upper concentration value of a nonionic surface-active substance is determined below which no further decrease in desorption of the emulsifier bound to an anion exchanger takes place,
- the aqueous phase is adjusted to a concentration of nonionic surface-active agent of between the upper concentration value determined in this way and a lower concentration which is still effective for preventing coagulation of the polymer particles,
- the aqueous phase adjusted in this way is brought into contact with an anionic exchanger resin in order to effect the adsorption of the emulsifier onto the exchanger resin, and
- the emulsifier is liberated from the exchanger resin.

The suitable concentration of nonionic surface-active agent is dependent on the type of polymer, on the surface-active agent and on any other substances present in the aqueous phase. It is therefore advisable to determine the suitable concentration limits of the nonionic surface-active agent for each waste water to be treated. A maximum concentration of 10 ppm, in most cases a concentration in the range from 5 to 0.1 ppm, is usually sufficient.

Since - as mentioned above - the waste water to be treated in accordance with the invention is preferably slightly contaminated, it is sensible to add only sufficient auxiliary chemicals to the waste water as is necessary in order to avoid causing fresh contamination for the further work-up of the waste water. If, on the other hand, it is desired to avoid the respective determination of the limit values in industrial practice, where mixtures of different waste water may have to be worked up together, a mean value of about 3 ppm can generally be used without problems.

A further advantage of the use of small amounts of nonionic surface-active agent, besides avoiding unnecessary costs, is also the suppression of foam, which can be very troublesome on an industrial scale and in some cases requires further contamination of the waste water with foam suppressors.

In the preparation of fluoropolymers, such as polytetrafluoroethylene, fluorinated thermoplastics and fluorinated elastomers, the polymers are separated off by coagulation, which is carried out mechanically with high shear ratios or chemically by precipitation with mineral acids or inorganic salts. The coagulated fluoropolymers are usually agglomerated and washed with water. Relatively large amounts of process waste water thus arise, namely usually from about 5 to 10 tonnes of waste water per tonne of fluoropolymer. In these process steps, the majority of the fluorinated emulsifier is washed out and is thus present in the waste water. The concentration is usually a few millimoles per liter, corresponding to approximately 1000 ppm. Besides the constituents already mentioned above, the waste water furthermore contains chemicals from the polymerization, such as initiators and buffers, which are present in approximately the same order of magnitude as the emulsifier, and very small amounts of fluoropolymer latex particles which have not been coagulated. The proportion of these latex particles in the waste water is usually less than 0.5% by weight.

It has already been mentioned that the preparation of fluorinated emulsifiers is associated with considerable effort, especially since these substances have to be employed in high purity.

Furthermore, these emulsifiers have poor biodegradability, and therefore the most complete removal possible from the waste water appears necessary. The process according to the

invention allows virtually quantitative recovery, even from the slightly contaminated types of waste water defined above.

A further advantage of the low concentrations of nonionic surface-active agent is the more effective separation of the latex particles from the anion-exchanged waste water. These particles are advantageously coagulated with small amounts of organic flocculating agents, it having been found that the amount of flocculating agent required increases with increasing concentration of nonionic surface-active agent. The resultant fluoropolymers, which are now contaminated with small amounts of surface-active agent and flocculating agent, can be used in building materials and therefore do not have to be subjected to complex work-up or disposed of to landfill.

Suitable nonionic surface-active agents are the commercially available oxyethylates and oxypropylates of organic hydroxyl compounds, preference being given to non-aromatic oxyalkylates for environmental protection reasons. Preference is therefore given to oxyethylates of long-chain alcohols.

Organic flocculating agents are described, for example, in *Encycl. Polym. Sci. Engng.*, Wiley Interscience, New York 7, 211 (1987).

The organic flocculating agents are advantageously cationic products, for example polydiallyldimethylammonium chloride.

Cationic surfactants, such as, for example, didecyldimethylammonium chloride, can likewise be used for precipitation of the nonionic stabilized latex particles. However, their use on an industrial scale is problematic since charge reversal of the particles to give cationically stabilized latex particles can take place preferentially if the precipitation is carried out incorrectly. This considerably reduces the degree of precipitation.

The invention is explained in greater detail in the examples below.

## Examples

In the examples below, waste water of mechanically coagulated polymer dispersions comprising about 90% by weight of the perfluorooctanoic acid employed in the polymerization as well as latex particles were employed. They are not diluted with washing water from the agglomerated resins. Waste water from the polymerization of tetrafluoroethylene with ethylene, polyfluoro(n-propyl vinyl) ether, hexafluoropropene and a terpolymer of tetrafluoroethylene, hexafluoropropene and vinylidene fluoride, and mixtures of such waste water, was investigated. Since it was found that waste water of said terpolymers and of the copolymers of tetrafluoroethylene and said ether and of ethylene tended to result in blockage of the exchanger column, this waste water was investigated in greater detail.

The dimensions of the anion exchanger column were as follows: height 5 cm, diameter 4 cm, fill amount 500 ml, flow rate from 0.5 to 1 l/h, working procedure: from top to bottom. A commercially available, strongly basic anion exchanger @AMPERLITE IRA 402, capacity 1.2 mmol/ml, was employed.

Blockage of the column was noted by monitoring the flow rate under a constant hydrostatic pressure. The experiments were carried out until the perfluorooctanoic acid appeared. A typical experiment on a laboratory scale required an amount of up to 150 l. The flow rate was determined at the beginning and at the end by weighing the exchanged waste water for a given time. A decrease in the flow rate of < 20% at the end of the experiment was regarded as acceptable. At the beginning of the experiment, the anion exchanger resin was in the OH<sup>-</sup> form. The determination limit for the perfluorooctanoic acid was 5 ppm.

### Example 1

A process waste water ("polymerization liquor") from the polymerization of the terpolymer of tetrafluoroethylene, hexafluoropropene and vinylidene fluoride containing 0.3% by weight of polymer latex particles and 0.1% by weight of perfluorooctanoic acid was employed. A commercially available p-octylphenol oxyethylate @TRITON X 100 (Rohm & Haas, CAS No. 9002-93-1) was employed.

**Table 1**

Concentration of @TRITON [ppm]	50	10	3	0.3
Total amount of waste water passed through [l]	125	150	150	150
Flow rate [l/h] at the beginning at the end	1.0 0.9	1.0 0.95	1.0 1.0	1.0 1.0
Concentration of perfluorooctanoic acid [ppm] after				
50 l	< 5	< 5	< 5	< 5
100 l	32	20	< 5	< 5
125 l	> 100	17	11	7
150 l	-	> 100	> 100	> 100

The perfluorooctanoic acid concentrations close to break-through exhibit "run-out" at relatively high concentrations of the nonionic surface-active agent. The nominal ion exchange capacity appears to be reduced at relatively high concentrations of nonionic surface-active agent.

**Example 2**

Example 1 is repeated with the modification that the nonionic surface-active agent employed was a commercially available fatty alcohol polyglycol ether @GENAPOL X 080 (Hoechst AG).

**Table 2**

Concentration of @GENAPOL [ppm]	300	30	3	0.3
Total amount of waste water passed through [l]	125	125	150	150
Flow rate [l/h] at the beginning at the end	1.0 0.9	1.0 0.9	1.0 0.95	1.0 0.95
Concentration of perfluorooctanoic acid [ppm] after				
50 l	< 5	< 5	< 5	< 5
100 l	23	18	< 5	< 5
125 l	> 100	> 100	12	8
150 l	-	-	> 100	> 100

**Example 3**

Example 2 was repeated, but a process water ("polymerization liquor") from the polymerization of a copolymer of tetrafluoroethylene with perfluoro(n-propyl vinyl) ether containing 0.1% by weight of perfluorooctanoic acid and 0.4% by weight of polymer latex particles was employed.

**Table 3**

Concentration of @GENAPOL [ppm]	30	3	0.3
Total amount of waste water passed through [l]	150	150	150
Flow rate [l/h] at the beginning at the end	1.0 0.9	1.0 1.0	1.0 0.9
Concentration of perfluorooctanoic acid [ppm] after			
50 l	< 5	< 5	< 5
100 l	< 5	< 5	< 5
125 l	15	7	9
150 l	> 100	> 100	> 100

**Example 4**

Example 2 was repeated, but a process water ("polymerization liquor") from the polymerization of a copolymer of tetrafluoroethylene with ethylene containing 0.2% by weight of perfluorooctanoic acid and 0.6% by weight of polymer latex particles was employed.

**Table 4**

Concentration of @GENAPOL [ppm]	30	3	0.3
Total amount of waste water passed through [l]	75	75	75
Flow rate [l/h] at the beginning at the end	0.5 0.45	0.5 0.45	0.5 0.45
Concentration of perfluorooctanoic acid [ppm] after			
50 l	< 5	< 5	< 5
100 l	< 5	< 5	< 5
125 l	35	12	10
150 l	> 100	> 100	> 100

**Example 5**

The types of waste water indicated in Tables 5 and 6 were treated with the commercially available organic flocculating agent @MAGNOFLOC 1697 (polydiallyldimethylammonium chloride, Allied Colloids Company). The minimum concentration of the flocculating agent for quantitative precipitation of the latex particles was determined by titration. A 0.1% strength by weight solution of the flocculating agent was added dropwise to the exchanger eluate with gentle stirring. The latex particles are precipitated virtually instantaneously and settle very quickly. The

dropwise addition is terminated when no further precipitate is observed. The results are shown in the following table.

**Table 5:** PFOA-free process waste water (PFOA concentration < 5 ppm)

Waste water from	Example 1			Example 3			Example 4		
®GENAPOL concentration [ppm]	300	30	3	300	30	3	300	30	3
Minimum concentration of flocculating agent [ppm]	23	3.5	2.6	27	3.0	2.7	35	9	7.7

5

**Table 6:** Untreated process waste water (PFOA concentration about 1000 ppm)

Waste water from	Example 1			Example 3			Example 4		
®GENAPOL concentration [ppm]	-	3	30	-	3	30	-	3	30
Minimum concentration of flocculating agent [ppm]	4.6	6	10	8.3	10	15	8.0	10	13

## Patent claims

1. A process for the recovery of fluorinated emulsifiers from an aqueous phase which, besides the emulsifier, contains small amounts of fluoropolymer particles, wherein
  - an upper concentration value of a nonionic surface-active substance is determined below which no further decrease in desorption of the emulsifier bound to an anion exchanger takes place,
  - the aqueous phase is adjusted to a concentration of nonionic surface-active agent of between the upper concentration value determined in this way and a lower concentration which is still effective for preventing coagulation of the polymer particles,
  - the aqueous phase adjusted in this way is brought into contact with an anionic exchanger resin in order to effect the adsorption of the emulsifier onto the exchanger resin, and
  - the emulsifier is liberated from the exchanger resin.
2. A process for the recovery of fluorinated emulsifiers from an aqueous phase which, besides the emulsifier, contains small amounts of fluoropolymer particles, wherein the aqueous phase is adjusted to a concentration of nonionic surface-active agent of between 10 ppm and a lower concentration which is still effective for preventing coagulation of the polymer particles, the aqueous phase adjusted in this way is brought into contact with an anionic exchanger resin in order to effect the adsorption of the emulsifier onto the exchanger resin, and the emulsifier is liberated from the exchanger resin.
3. The process as claimed in claim 2, where the concentration of the nonionic surface-active agent is from 5 to 0.1 ppm.
4. The process as claimed in one or more of the preceding claims, wherein the nonionic surface-active agent is non-aromatic.
5. The process as claimed in one or more of the preceding claims, where the nonionic surface-active agent is a fatty alcohol oxyethylate.

6. The process as claimed in one or more of the preceding claims, where an effective amount of an organic flocculating agent is added to the aqueous phase in order to precipitate essentially all the fluoropolymer particles.
- 5 7. The process as claimed in claim 6, wherein the flocculating agent is an organic cationic flocculating agent.
8. The process as claimed in claim 6 or 7, wherein the flocculating agent is added to the untreated or treated aqueous phase.

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**§1.56 Duty to disclose information material to patentability.**

(a) A patent by its very nature is affected with a public interest. The public interest is best served, and the most effective patent examination occurs when, at the time an application is being examined, the Office is aware of and evaluates the teachings of all information material to patentability. Each individual associated with the filing and prosecution of a patent application has a duty of candor and good faith in dealing with the Office, which includes a duty to disclose to the Office all information known to that individual to be material to patentability as defined in this section. The duty to disclose information exists with respect to each pending claim until the claim is cancelled or withdrawn from consideration, or the application becomes abandoned. Information material to the patentability of a claim that is cancelled or withdrawn from consideration need not be submitted if the information is not material to the patentability of any claim remaining under consideration in the application. There is no duty to submit information which is not material to the patentability of any existing claim. The duty to disclose all information known to be material to patentability is deemed to be satisfied if all information known to be material to patentability of any claim issued in a patent was cited by the Office or submitted to the Office in the manner prescribed by §§ 1.97(b)-(d) and 1.98. However, no patent will be granted on an application in connection with which fraud on the Office was practiced or attempted or the duty of disclosure was violated through bad faith or intentional misconduct. The Office encourages applicants to carefully examine:

- (1) prior art cited in search reports of a foreign patent office in a counterpart application, and
- (2) the closest information over which individuals associated with the filing or prosecution of a patent application believe any pending claim patentably defines, to make sure that any material information contained therein is disclosed to the Office.

(b) Under this section, information is material to patentability when it is not cumulative to information already of record or being made of record in the application, and

- (1) It establishes, by itself or in combination with other information, a prima facie case of unpatentability of a claim; or
- (2) It refutes, or is inconsistent with, a position the applicant takes in:
  - (i) Opposing an argument of unpatentability relied on by the Office, or
  - (ii) Asserting an argument of patentability.

A prima facie case of unpatentability is established when the information compels a conclusion that a claim is unpatentable under the preponderance of evidence, burden-of-proof standard, giving each term in the claim its broadest reasonable construction consistent with the specification, and before any consideration is given to evidence which may be submitted in an attempt to establish a contrary conclusion of patentability.

(c) Individuals associated with the filing or prosecution of a patent application within the meaning of this section are:

- (1) Each inventor named in the application;
- (2) Each attorney or agent who prepares or prosecutes the application; and
- (3) Every other person who is substantively involved in the preparation or prosecution of the application and who is associated with the inventor, with the assignee or with anyone to whom there is an obligation to assign the application.

(d) Individuals other than the attorney, agent or inventor may comply with this section by disclosing information to the attorney, agent, or inventor.

**DECLARATION, POWER OF ATTORNEY, AND PETITION**

I, a below named inventor, depose and say that: (1) my residence, citizenship, and mailing address are indicated below; (2) I have reviewed and understand the contents of my patent application, including the claims, as amended by any amendment specifically referred to herein, which is identified as PCT International Patent Application Serial No. PCT/EP00/06556 filed July 11, 2000, bearing Attorney Docket Number 56603WO004; (3) I believe that the other below named inventors and I are the original, first, and joint inventors of the invention or discovery in

**PROCESS FOR THE RECOVERY OF FLUORINATED EMULSIFIERS FROM AQUEOUS PHASES**

described and claimed therein and for which a patent is sought; and (4) I hereby acknowledge my duty to disclose to the Patent and Trademark Office all information known to me to be material to the patentability as defined in Title 37, Code of Federal Regulations, §1.56\*, and that no application for patent or inventor's certificate on this invention or discovery has been filed by us or our legal representatives or assigns in any country foreign to the United States of America except Application No. filed upon which we hereby claim foreign priority benefits under Title 35, United States Code Section 119.

I hereby appoint Gregory D. Allen (Reg. No. 35,048), Alan Ball (Reg. No. 42,286), Scott A. Bardell (Reg. No. 39,594), Carolyn A. Bates (Reg. No. 27,853), Bruce E. Black (Reg. No. 41,622), Colene E. H. Blank (Reg. No. 41,056), Jennie G. Boeder (Reg. No. 28,952), William J. Bond (Reg. No. 32,400), Arthur J. Brady (Reg. No. 42,356), Stephen W. Buckingham (Reg. No. 30,035), John A. Burtis (Reg. No. 39,924), Melissa E. Buss (Reg. No. 47,465), Gerald F. Chernivec (Reg. No. 26,537), James D. Christoff (Reg. No. 31,492), Philip Y. Dahl (Reg. No. 36,115), Janice L. Dowdall (Reg. No. 31,201), Lisa M. Fagan (Reg. No. 40,601), Carolyn A. Fischer (Reg. No. 39,091), Yen T. Florczak (Reg. No. 45,163), Darla P. Fonseca (Reg. No. 31,783), Melanie G. Gover (Reg. No. 41,793), Christopher D. Gram, (Reg. No. 43,643), Gary L. Griswold (Reg. No. 25,396), Doreen S. L. Gwin (Reg. No. 35,580), Michael A. Hakamaki (Reg. No. 40,011), Karl G. Hanson (Reg. No. 32,900), Dean M. Harts (Reg. No. 47,634), Néstor F. Ho (Reg. No. 39,460), Rudolph P. Hofmann, Jr. (Reg. No. 38,187), Robert W. Hoke (Reg. No. 29,226), MarySusan Howard (Reg. No. 38,729), Stephen C. Jensen (Reg. No. 35,207), Robert H. Jordan (Reg. No. 31,973), Harold C. Knecht III (Reg. No. 35,576), Kent S. Kokko (Reg. No. 33,931), Andrew J. Leon (Reg. No. 46,869), James V. Lilly (Reg. No. 27,817), Douglas B. Little (Reg. No. 28,439), Eloise J. Maki (Reg. No. 33,418), Michelle M. Michel (Reg. No. 33,968), William D. Miller (Reg. No. 37,988), Peter L. Olson (Reg. No. 35,308), Daniel R. Pastirik (Reg. No. 33,025), David B. Patchett (Reg. No. 39,326), Robert J. Pechman (Reg. No. 45,002), Carolyn V. Peters (Reg. No. 33,271), Scott R. Pribnow (Reg. No. 43,869), Ted K. Ringsred (Reg. No. 35,658), Steven E. Skolnick (Reg. No. 33,789), Robert W. Sprague (Reg. No. 30,497), Brian E. Szymanski (Reg. No. 39,523), James J. Trussell (Reg. No. 37,251), Lucy C. Weiss (Reg. No. 32,834), Bradford B. Wright (Reg. No. 34,459), and Kimberly S. Zillig (Reg. No. 46,346) my attorneys and/or agents with full powers (including the powers of appointment, substitution, and revocation) to prosecute this application and any division, continuation, continuation-in-part, reexamination, or reissue thereof, and to transact all business in the Patent and Trademark Office connected therewith; the mailing address and the telephone number of the above-mentioned attorneys and/or agents are

Attention: James V. Lilly  
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The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1-00

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DEX

3-00

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